Application No. 10/624,944

Reply to Office Action of November 29, 2006

REMARKS

Claims 1-12 are pending and stand rejected. Reconsideration of the application is respectfully requested in view of the Remarks below.

Rejections under 35 U.S.C. § 103(a)

Claims 1-7 and 9-12 stand rejected as obvious over Harada et al (U.S. Published Application No. 2002/0090335) in view of Guo et al (U.S. Patent No. 6,827,916) and Vita et al (U.S. Patent No. 2,985,506).

According to the Examiner, Harada teaches that the hydroxide ion is added to reaction (paragraphs 68 and 69) in excess (paragraph 69) and from this teaching, "it would be obvious that the pH is maintained at 14 and that because an excess of hydroxides is present, for all intents and purposes, the amount of OH is constant."

Applicants respectfully disagree with the Examiner's position and request reconsideration in view of the below remarks.

The cited references do not teach or suggest "maintaining the reaction mixture at a constant OH concentration"

Claim 1 recites, inter alia, "maintaining the reaction mixture at a constant OH concentration" which is purportedly disclosed in Harada. Harada, however, merely states at paragraphs 68 and 69 that the titanium hydroxide colloid employed is produced by neutralizing an aqueous titanium salt solution with an aqueous alkali solution (e.g. an aqueous solution of a base such as sodium hydroxide, potassium hydroxide, ammonia or the like). These two paragraphs describe how to obtain the titanium hydroxide colloid and explain that the hydroxide ion is used for the purpose of neutralization.

in the art to conclude that the amount of OH is kept constant based on this disclosure. Such an excess of base is used merely to neutralize an excess of acid added for preventing the hydrolysis of titanium when formulating a titanium salt solution, which is well known in the art.

To the contrary, claim 1 of the present application recites that the reaction mixture is maintained at constant OH concentration. This limitation is not disclosed or suggested by Harada, including paragraphs 68 and 69 cited by the Examiner. As the Examiner implicitly admits, this limitation is also not taught by the remaining references cited by instant rejection.

The "optimization" in Vita is directed to a different purpose and hence would not lead a person of ordinary skill to the instantly claimed ranges

The Examiner states that Vita teaches the optimization of feed solutions to obtain a desired particle size of barium titanate. According to the examiner's reasoning, rate of flow and concentration of the instant claims can be obtained based the teaching of Vita, which teaches varying such parameters to control the crystal form and particle size of the barium titanate. Applicants respectfully disagree with this assertion.

According to the present invention, the purpose of altering the concentrations and flow ratio of reaction solutions is to control the degree of supersaturation of the product and in turn to control the particle size and the Ba/Ti ratio of the product. In contrast, Vita seeks merely to obtain a specific crystal form. Therefore, the examiner's opinion that discovering an optimum value or a result effective variable in view of Vita involves only routine skill in the art is incorrect, as any opitimization efforts in Vita is directed toward a different purpose. Within a certain range, any alterations according to the present invention result in the production of barium titanates, which can find different uses. In view of the above, it would be unobvious to optimize the flow rates of the reactants to obtain the presently claimed process, including the alkali solution, of Harada et al.

More particularly, the reaction according to the present invention is carried out in accordance with the principle of one-step process and macroscopically, the aqueous solution of titanium and barium is reacted with an excess of OH in one step -- which is instantaneous and rapid -- to obtain barium titanate powders as described in the present invention. The thermodynamic and kinetic mechanisms of this process is described in more detail in Attachment 1 (Chinese J Chem Eng, Vol 13, No 2(2005), p. 225-233) and Attachment 2 (Chinese J Chem Eng, Vol 14, No 5(2006), p. 642-648).

According to these attachments, the reaction according to the present invention is carried out in a gel dissolution-precipitation reaction mechanism illustrated in Figure 1.

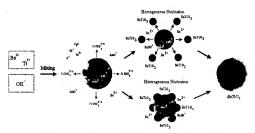


Figure 1 Schematic sketch of the gel dissolution-precipitation reaction mechanism

As shown in Figure 1, in the first stage of the reaction according to the present invention, the solution containing Ba²⁺ and Ti⁴⁺ is contacted with a strong alkali solution containing a large number of OH ions to form a porous sol system containing Ti-O anionic complex as the skeleton and a great deal of heat is released, since the aqueous titanium tetrachloride solution is easily hydrolysable to form a network-like Ti-O anionic complex and Ba ions are not easily complexible. This stage of reaction can be illustrated by the following formula (1) and is referred to as in-situ reactive transformation. At the same time,

there are a large number of barium ions and other ions adsorbed on the porous sol and in terms of reaction time, this kind of reaction is instantaneous. In the second stage of the reaction according to the present invention, BaTiO₃(s) is formed from the Ti-rich sol formed in the first stage and Ba²⁺(or BaOH⁺) ions adsorbed on the sol due to the thermal induction and the impingement between the nucleophilic OH⁻ and Ti-O anionic complex, with some heat being released. This stage of reaction is illustrated by the following formula (2) and is referred to as dissolution-precipitation and constitutes the rate determining step of the overall reaction. Due to this, the formation of BaTiO₃(s) is self-catalyzed to a certain degree and thus barium titanate is formed in the solution in a large amount, which results in a higher degree of supersaturation. After this, nucleation and growth occur and finally crystalline barium titanate particles having a specific morphology are obtained.

$$Ti^{4+}_{(aq)} + nOH^{-}_{(aq)} \rightarrow Ti_{Sol} + mH_2O$$
 (1)

$$Ti_{Sol} + Ba^{2+}(BaOH^+)_{(aq)} + uOH^-_{(aq)} \rightarrow BaTiO_{3(s)} + vH_2O_{(l)}$$
 (2)

The above study on the kinetics made by the present inventors illustrates why the conventional process needs 6 hours of aging to obtain barium titanate particles having a higher purity.

Microscopically, since the reaction according to the above formula (2) is the rate determining step, the concentration of barium salt in the system must be enhanced so as to obtain finer barium titanate powders. Only by this, a large amount of barium titanate can be formed in a short time, which in turn results in a higher degree of supersaturation. Finally, finer barium titanate particles are formed in the Higee reactor and are discharged. In the meantime, such a mechanism demonstrates that it is necessary for the concentration of OH to be maintained constant(cf. the principle illustrated by formula (1)). Vita et al mentions in lines 55 to 60, column 2 that by regulating the rate of flow and the concentration of the solution and by regulating the rate of flow of the combustion gas it becomes possible to

control the temperature of the reaction, the crystal form and the particle size of the formed barium titanate crystals. It is obvious that the reaction disclosed in Vita et al belongs to a solid reaction and the purpose of regulating the rate of flow is to regulate the temperature of reaction and that of regulating the concentration of the solution is to control the particle size. It should be pointed out that in the present invention, a smaller particle size can be obtained by regulating the concentration of the reaction solutions, which neither represents the optimization in the sense of chemistry and engineering nor means that no barium titanate products can be obtained at other concentrations, and to the contrary, only demonstrates that barium titanate powders having a different size and finding different uses can be obtained by regulating the concentration of the reactants.

The Higee reactor in Guo is for a different purpose; there is no motivation to employ it in the instant process absent impermissible hindsight

The Higee reactor in Guo has a different purpose and is based on a different principle compared with the use of Higee reactor in the present invention. More particularly, a Higee reactor is used in Guo et al to improve the gas/liquid mass transfer velocity of the reactants and in turn increase the reaction efficiency(which is derived from the absorption restriction of CO₂). In the present invention, a Higee reactor is used to instantaneously improve the microscopic mixing degree of the reactants(liquid/liquid) so as to realize the above-mentioned reaction process, that is to say, rapidly form finer barium titanate powders. In this case, the reaction instantaneously occurs in the extremely small micro-elements produced by the action of the Higee reactor. It is obvious that the invention of Guo et al employs a different action of the Higee reactor compared with the present invention and a person skilled in the art cannot conceive the present invention by simply combining Harada et al and Guo et al. Only upon the impermissible use of hindsight would one of ordinary skill be motivated to employ the Higee reactor in the presently claimed process.

More particularly, although a Higee reactor is favorable to the reaction step of forming barium titanate starting particles in Harada et al(lines 1 to 7, Paragraph 67), such a step is only the first step for preparing spherical tetragonal barium titanate particles according to Harada et al. After this, the formed barium titanate starting particles need to be subjected to hydrothermal treatment so as to obtain some physical features similar to those of the products formed in one step in the Higee reactor according to the present invention. It is obvious that the thermal treatment cannot be completed in the Higee reactor, since the latter cannot be used for thermal treatment. Therefore, it is impossible for a person skilled in the art to achieve the overall object of Harada et al in the Higee reactor disclosed in Guo et al

In view of the above, it is impossible for a person skilled in the art to conceive the technical solution of the present invention by simply combining the reaction of Harada et al with the optimization conceptation of Vita et al and the Higee reactor of Guo et al.

Claim 2 stands rejected as obvious over Harada in view of Guo, Vita and Kawamoto (U.S. Published Application No. 2003/0022784). The rejection relies upon Kawamoto to teach a process for producing barium titanate (Paragraph 0003, lines 1 to 3) wherein the Ba/Ti molar ratio is 1.16 (Paragraph 0022, lines 1 to 5) for the purpose of producing the desired stoichiometry of barium and titanium (Paragraph 0016, lines 10 to 13). Claim 5 stands rejected as obvious over Harada in view of Guo, Vita and Kerchner (U.S. Patent No. 6,129,903). The Examiner relies upon Kerchner to teach a process for producing barium titanate powders (col. 1, lines 13 to 15) wherein the addition of the barium source increases the pH of the reaction mixture to about 13 (col. 5, lines 22 to 25) for the purpose of promoting the reaction (col. 5, lines 23 to 25).

Applicants submit that claims 2 and 5 are unobvious due at least to the reasons set forth above. Furthermore, with respect to the Ba/Ti molar ratio of more than 1, the present

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invention cannot be simply compared with Kawamoto et al. since these two inventions are based on different mechanisms and in addition, the action of Ba ions in the reaction process is different. In fact, the amount of barium added is related to the reaction kinetics and according to the above study on the reaction kinetics, an excess of barium is favorable to the reaction between barium and Ti-O anionic complex, which in turn results in finer barium titanate product having a stroichiometric ratio. With respect to the teaching of pH in Kerchner et al. Kerchner et al mentions in the process for producing barium titanate powders that the addition of the barium source increases the pH of the reaction mixture, which is obviously determined by a specific reaction mechanism. However, the further increase in the amount of barium added in the present invention is insufficient to alter the pH of the reaction system, since on one hand barium chloride added cannot provide OH and on the other hand the high pH possessed by the reaction system is established by adding a large amount of alkali solution. That is to say, Kerchner et al adopts a measure clearly different from the present invention to maintain the pH.

Based on the above reasons, the applicant requests that the obviousness rejections be withdrawn. If an interview is deemed to advance prosecution, the Examiner is respectfully requested to contact the undersigned at the number below.

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